Magneto-Resistant Effect of MgO-Based Double-Barrier Magnetic Tunnel Junctions

著者	姜 麗仙
号	54
学位授与機関	Tohoku University
学位授与番号	工博第4284号
URL	http://hdl.handle.net/10097/61561

	じゃん　りーしぇん
氏　　　　　名	姜　　麗仙
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指　導　教　員	東北大学教授　安藤　康夫
論文審査委員	主査　東北大学教授　佐久間　昭正　東北大学教授　北上　修

論文内容要旨

Introduction

The spin is a basic feature of the electron. Adding and using the spin degree of freedom offer interesting possibilities for the development of spintronics device. The tunneling of electron in Ferromagnetic electrode/Insulator/Ferromagnetic electrode junctions (called as magnetic tunnel junction) shows a resistance change depending on the magnetization alignment of two ferromagnetic electrode layers. This effect is called as tunneling magnetic resistance (TMR) effect. Recently, developed spintronics devices such as magnetic random access memory (MRAM), magnetic sensor and spin-transistor require high TMR ratio in magnetic tunnel junction (MTJ). The TMR ratio increase from below 100% in MTJ with Al-O_x as barrier to about 200% in MTJs with MgO (100) barrier. For application of MRAM, the TMR ratio is 200% and R-H loop of MTJ with spin-valve type are enough. However, the application of new logic device and high sensitive sensor require over 1000% high TMR ratio. Theoretically, the double-barrier magnetic tunnel junctions (DBMTJs), which can be used for three-terminal device such as spin transistor, have a possibility to enhance the TMR ratio due to quantum well (QW) states created in the middle free ferromagnetic layer. Additionally, Coulomb blockade (CB) effect and spin accumulation effect occurs in the middle layer strongly depending on the DBMTJs stacking structure. However, experimentally, there is no report about huge TMR ratio induced by quantum well (QW) resonant effect and Coulomb blockade (CB) effect in the DBMTJs structure. One of the reasons of low TMR ratio is quality of interfacial structure of the DBMTJs. Therefore, in order to create quantum effect, it is necessary to fabricate the high-quality DFBMTJs with thin middle layer. The purpose of this study is fabricate high quality MgO-based DBMTJs with various thickness $Fe_{50}Co_{50}$ or $Fe_{40}Co_{40}B_{20}$ as middle free layer to obtain high TMR ratio and quantum effect (QW resonant tunneling and CB effect).

Experimental method in chapter 2

The multilayer deposited by magnetron sputtering system was patterned to DBMTJs using micro-fabrication. The MgO barrier was deposited by RF-sputtering from MgO target. The size of junction was 20×10, 40×20, and 80×40 μm^2. After the fabrication, the DBMTJs were annealed at temperature ranging from 250 to 375°C. The magnetic property of middle ferromagnetic layer was measured by superconductive quantum interface (SQUID) magnetometer. The structure of the multilayer was observed by transmission electron microscopy (TEM). The magnetic transport property was measured using a standard four-probe method.

Experimental results in chapter 3

In the MgO-based DBMTJ with $Fe_{50}Co_{50}$ middle free layer of 1.5 nm in thickness, the large negative TMR ratio of -46% (Fig. 1) was obtained at room temperature (RT). The bias voltage dependence of TMR showed very unique behavior. The TMR ratio was negative at negative voltage, while the TMR ratio was positive at positive voltage. The same behavior was also observed in the DBMTJ with $Fe_{50}Co_{50}$ middle free layer of 1.7 and 2.3 nm. The origin of negative TMR might be attributed to the formation of FeCo-O_x from MgO barrier at the interface.

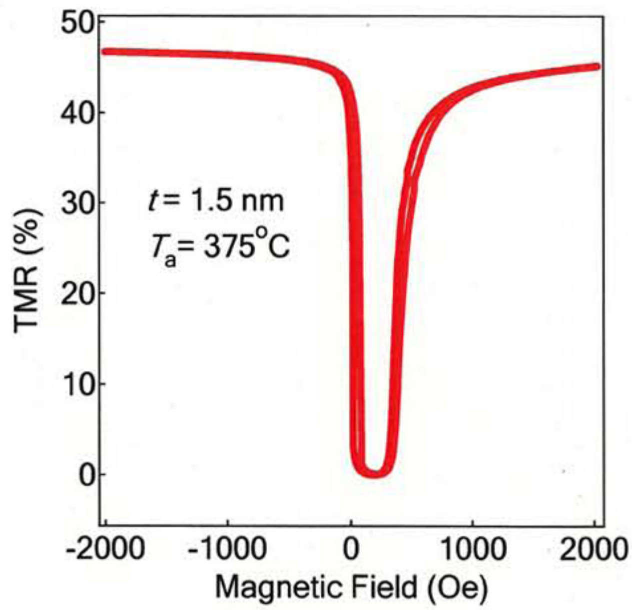

Fig. 1. The *R-H* curve of the DBMTJ with 1.5 nm $Fe_{50}Co_{50}$ as middle layer at a bias voltage -500 mV. at RT.

Experimental results in chapter 4

In the case of the MgO-based DBMTJ with thin $Fe_{40}Co_{40}B_{20}$ middle free layer, the TMR ratio depended on the thickness of the middle layer. The TMR ratio of 1056% was obtained in the DBMTJ system with very thin middle layer of 1.2 nm in thickness (Fig. 2), which is the highest value at RT reported to date. Moreover, the DBMTJ with spin-valve type exhibited very sharp magnetization switching as well as the low resistance-area (RA) product of 2×10^4 $\Omega\mu m^2$ although the thickness of each MgO barrier was 2.5 nm. These fascinating magnetotransport properties in the DBMTJs are attracted from the viewpoint of spintronics applications. However, when the thickness is further increased to 1.5 nm, the TMR ratio drastically decreased to 80% and the RA also dramatically increased, which indicates the high TMR ratio is obtained at very thin middle layer of the DBMTJs. When focusing *J-V* characteristics, the clear stair cases were observed in the *J-V* curves with middle layer thickness of 0.8, 1.0 and 1.2 nm. As shown in the Fig. 3, the clear staircase was observed at RT in the *J-V* curves for the DBMTJs with the middle layer thickness of 1.2 nm. The threshold voltage for parallel and antiparallel configuration was 55 mV and 100 mV, respectively. In the case of 0.8 nm, a clear staircase with a large threshold voltage of 100 mV (200 mV) for parallel (antiparallel) magnetization configuration was observed at RT. This is the first time that such threshold voltage was observed at RT. The bias voltage dependence of TMR ratio (*TMR-V*) estimated from the *I-V* curves showed sharp peaks in both positive and negative voltage. When the thickness of the middle layer further increased to 1.5 nm, the junction showed the normal *I-V* curves and large RA. These attractive phenomena in the DBMTJ with

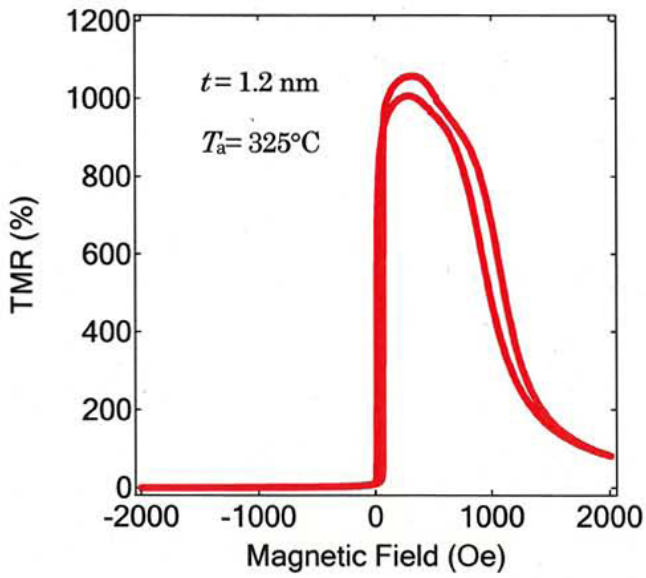

Fig. 2. The *R-H* curve of the DBMTJ with 1.2 nm $Fe_{40}Co_{40}B_{20}$ as middle layer at a bias voltage -70 mV at RT.

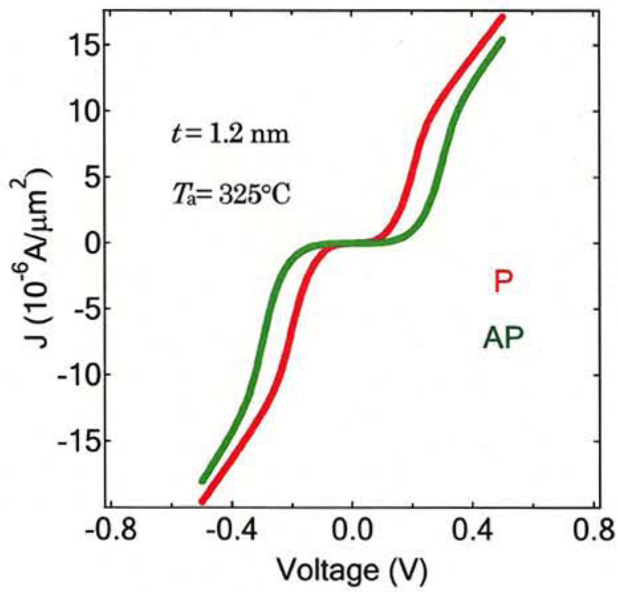

Fig. 3. Current density *J-V* curves for DBMTJ with 1.2 nm $Fe_{40}Co_{40}B_{20}$ as middle layer at RT.

below 1.2 nm middle free layer were discussed by conventional QW state resonant tunneling and CB effect. The interesting feature is that both theories can not perfectly be explained the magnetotransport properties. We can expect new physics of magnetotransport properties, which has a possibility to open new field of spintronics.

Conclusions

The largest TMR ratio of 1056% reported to now and low RA of 2×10^4 $\Omega\mu m^2$ was observed at RT in MgO-based DBMTJs with $Fe_{40}Co_{40}B_{20}$ as the middle free layer. The *R-H* loop in this DBMTJ with spin valve type show sharp magnetization switching. These wonderful features are very useful for application of MRAM and magnetic high sensitive sensor applications. In addition, it is the first time that the clear stair case with large voltage threshold in the *I-V* curve was obtained at RT. These features attract the researcher to realization of spin-transistor. And all these amazing experimental results can contribute more and more to development in spintronics research field.

論文審査結果の要旨

本論文では、MgO 障壁を有する二重強磁性トンネル接合における磁気抵抗効果を調べることを目的とした。第 1 章は序論である。本研究を記述する上で欠かせない、強磁性トンネル接合における磁気抵抗比の改善の推移、二重強磁性トンネル接合における量子井戸効果、共鳴トンネル効果、およびクーロンブロッケイド効果に関する記述を行った上で、研究の目的として、本研究の位置づけと研究目的について述べている。

第 2 章は実験方法で、二重強磁性トンネル接合の成膜、磁気抵抗効果評価用の素子微細加工に関して述べている。また、試料の熱処理方法、測定方法についても記述されている。

第 3 章は中間層として CoFe を用いた二重強磁性トンネル接合における磁気抵抗効果について記述している。中間層の膜厚を 1.5 nm とすることで-46%の大きな負の磁気抵抗効果が観測された。同様な現象は他の中間層の膜厚においても観測された。この結果は CoFe の表面が酸化することにより現れたものと解釈された。

第 4 章は中間層として CoFeB を用いた二重強磁性トンネル接合における磁気抵抗効果について記述している。中間層の膜厚が 1.2 nm の試料を 325℃で熱処理することにより、1056%の巨大な磁気抵抗効果を観測することに成功した。このときの磁気抵抗曲線は典型的なスピンバルブ型になっていることから、単に大きな磁気抵抗比のデモンストレーションという観点でなく、デバイスへの応用が可能な素子であることが併せて確認された。この試料の電流－電圧特性を測定すると、約数百 mV の閾電圧以上で電流が急激に立ち上がる、特異な曲線が得られた。またこの閾電圧値は強磁性体の磁化配列が平行のときよりも反平行のときの値が大きくなることがわかった。このような特異な電流－電圧曲線は、クーロンブロッケイド現象がおきているときに観測される現象と類似しているものの、スピン依存している点は従来のクーロンブロッケイドの理論では説明できない。一方、上記の巨大な磁気抵抗効果が観測されるときは素子の抵抗値が大きく減少することがわかっている。これは共鳴トンネルが生じていることを示唆するが、中間層の膜厚が 1.2 nm のときのみに共鳴トンネルが観測されることは、従来の理論では説明できない。これらのことから、今回観測された現象は新しい量子効果を考慮する必要があることが記述されている。

第 5 章は総括である。本研究の成果は、次世代の磁気メモリおよび高感度の磁気センサーの実現において大きく貢献するとともに、これまでにない新しい物理現象による磁気抵抗効果の発現によるスピントロニクス業界のイノベーションにつながる成果であり、応用物理学の発展に寄与するものであると期待される。

よって、本論文は博士(工学)の学位論文として合格と認める。